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Thermo-electric finite element analysis and characteristic of thermoelectric generator with intermetallic compound

Li-Ling Liao [a,b], Ming-Ji Dai [a], Chun-Kai Liu [a], Kuo-Ning Chiang [b,c,*]

[a] Electronic and Optoelectronics Research Laboratories, Industrial Technology Research Institute, HsinChu 300, Taiwan, ROC
[b] Dept. of Power Mechanical Engineering, National Tsing Hua University, HsinChu 300, Taiwan, ROC
[c] Advanced Packaging Research Center, National Tsing Hua University, HsinChu 300, Taiwan, ROC

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ABSTRACT

Energy recycling is an important research topic worldwide. Thermoelectric generators (TEGs) are used to recycle energy. TEGs are suitable for small and medium-sized applications and industrial waste heat harvesting in medium to high temperatures. Traditional lead-free soldering cannot be applied at temperatures above 200 °C. The joint may melt and fail at exceedingly high temperatures. Therefore, determining the suitable bonding material to apply during TEG assembly is necessary. Ni/Sn/Ag was adopted as the joint material for TEG packaging in this study. The joints were bonded and transformed into a full intermetallic compound (IMC) through solid–liquid interdiffusion bonding (SLID) and solid–solid interdiffusion reaction. The IMC has a high melting point and can be utilized at temperatures above 200 °C. The mechanical strength of the joints was examined through shear test at different thermal treatment times. Shear strength declined with the increase in thermal treatment time. The TEG was assembled with 12 pairs of thermoelectric (TE) pillars, and its performance was measured through the slope method. The TE characteristics do not significantly vary before and after thermal treatment. With the established finite element (FE) model, temperature and output power were estimated at specific temperature loading through thermo-electrical FE analysis.

1. Introduction

Energy recycling is an important research topic worldwide. Thermoelectric generators (TEGs) are utilized to recycle energy. The advantages of TEGs include convenience, high reliability, and environmental friendliness. Given its flexibility in structural design and size, the TEG is suitable for small and medium-sized applications as well as industrial waste heat harvesting in medium to high temperatures. The TEG has a solid-state heat engine fabricated with semiconductor materials. The working principle of the TEG is based on the Seebeck effect. Both sides of the thermoelectric (TE) pillar are maintained by the different junction temperatures, which result in an open-circuit electromotive force as shown in Fig. 1. The equation of the Seebeck effect is as follows [1]:

$$\alpha = \frac{V}{T_h - T_c} \tag{1}$$

where α is the Seebeck coefficient, V is the voltage produced, and T_h and T_c are the temperatures on the hot and cold sides, respectively.

The characteristic of the TE materials is determined by the figure of merit (ZT). The equation of ZT is as follows [1]:

$$Z = \frac{\alpha\sigma}{\kappa} \tag{2}$$

where Z is the figure of merit, σ is the electrical conductivity, and κ is the thermal conductivity.

The TEG consists of an array of p- and n-type TE materials bonded between two substrates as shown in Fig. 2 [2]. These materials are connected electrically in a series to form a chain of p–n junctions but are thermally connected in a parallel arrangement. The TE materials are attached to the substrates by soldering. Many studies investigated the interfacial behavior of TE materials and bonding joints. Lead-free Sn-based solders are widely utilized as the bonding material in TEGs. However, the solder may creep, crack, and even melt when the operating temperature is at or above 150 °C [3–5]. Nickel is commonly used as a diffusion barrier to prevent extensive interdiffusion and interfacial reactions. Nickel diffuses readily in bismuth telluride compounds at elevated temperatures during soldering or annealing [6,7]. The joint materials and diffusion barrier should be selected with caution because both should be able to sustain high temperatures. A thermal–electrical–mechanical analysis was conducted in the simulation phase to investigate the output voltage, output power, and thermal stress

* Corresponding author at: Dept. of Power Mechanical Engineering, National Tsing Hua University, HsinChu 300, Taiwan, ROC. Tel.: +886 3 5742925; fax: +886 3 5745377.
E-mail address: knchiang@pme.nthu.edu.tw (K.-N. Chiang).

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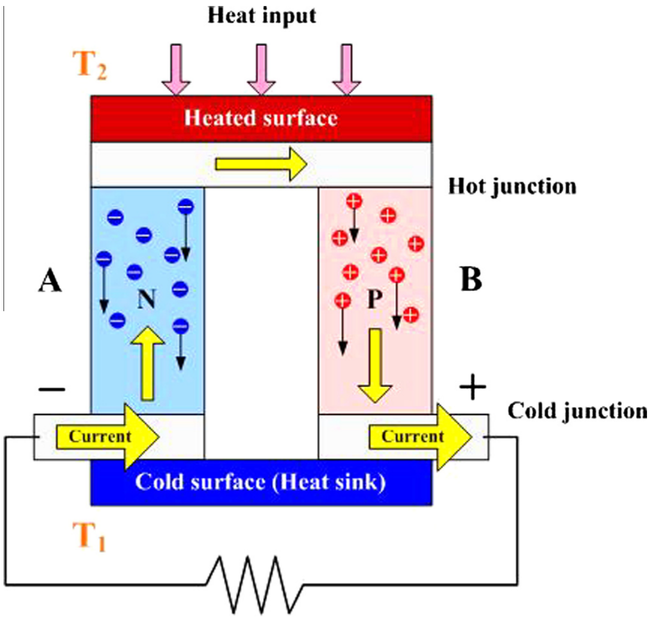

Fig. 1. Schematic diagram of the Seebeck effect.

of the TEG induced through a difference in temperature. The electrical and thermal simulation results were compared with the experimental results at different temperature conditions [8].

This research aims to investigate the use of intermetallic compounds (IMC) as a replacement for traditional solder materials in the interconnection joints. Ni/Sn/Ag was adopted as the joint material to assemble and form the TEG. After the bonding and thermal treatment processes, tin reacted with nickel and silver was entirely consumed to transform into a full IMC. The TEG with full IMC joints can be applied at temperatures above 200 °C because of the high melting point of the IMCs.

The composition and growth thickness of the IMCs were analyzed and examined through scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS). The mechanical strength of the IMCs was measured at different thermal treatment times through a shear test. An increase in thermal treatment time

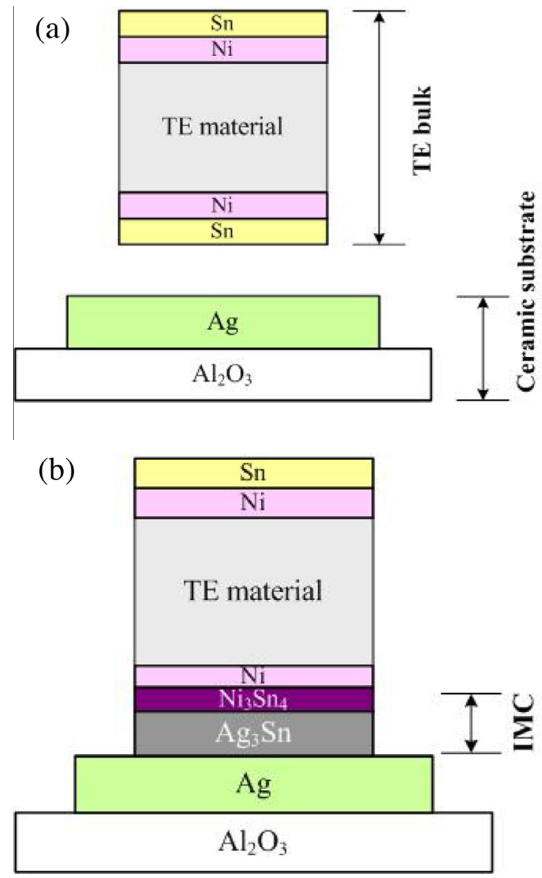

Fig. 3. (a) Schematic of the test sample and (b) tin was entirely consumed after reacting with nickel/silver to transform into a full IMC after bonding and thermal treatment process.

caused a decrease in shear strength. The fractured surface of the joints was observed and examined to understand structural strength after the shear test.

After TEG assembly, the TE characteristic of the TEG was measured through the slope method to obtain the Seebeck coefficient,

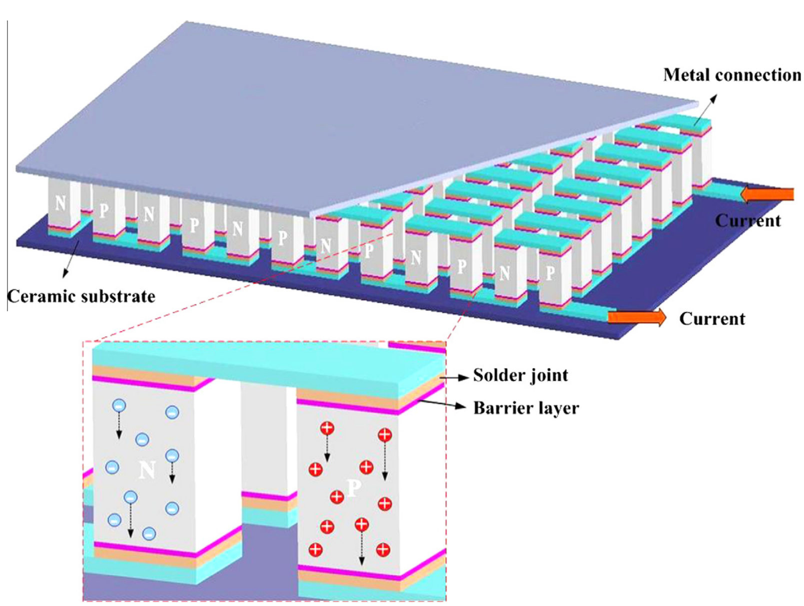

Fig. 2. Schematic diagram of the thermoelectric module.

resistivity, and thermal conductivity of the materials and the module. A 3D finite element (FE) model based on an actual TEG was constructed, and the thermo-electrical behavior was predicted through thermo-electrical FE analysis. Temperature distribution and opening voltage were estimated at different temperature loads.

2. Materials and methods

The TE material was purchased from a commercial vendor (Kryotherm, Russia) and was manufactured with bismuth, tellurium, antimony, and selenium at 99.999% purity. Both sides of the TE material were plated with 5 μm nickel and 10 μm tin. Nickel and tin functioned as the diffusion barrier and the bonding material, respectively. Silver (25 μm) with excellent electrical and thermal conductivity was sintered as the metal electrode on the ceramic substrate.

Fig. 3(a) shows a schematic of the test sample. Ni/Sn/Ag was utilized as the joint material, and the TE materials were bonded to the ceramic substrate through SLID bonding. The bonding conditions show that the bonding temperature is 280 °C, bonding force is 5 g, and holding time is 5 s. When the bonding temperature was higher than the melting point of tin, tin reacted with silver and nickel to form Ag_3Sn and Ni_3Sn_4 intermetallic compounds in the joints. Tin was entirely consumed after reacting with nickel/silver to transform into a full IMC through solid–solid interdiffusion reaction at thermal treatment temperature as shown in Fig. 3(b).

A shear tester (Dega 4000, England) was employed to conduct the shear test of the joints. Shear force was measured and calculated with the shear strength equation to obtain the shear strength of the joint. The sheared area of the joint is 1 × 1 (mm^2). The push knife of the shear tester was set 10 μm away from the electrode surface to confirm that the fracture behavior occurred from the Ag_3Sn IMCs. The joint fracture was observed and examined through SEM and EDS after the shear test. The shear strength of the joints can help us understand mechanical strength in the long-term operation.

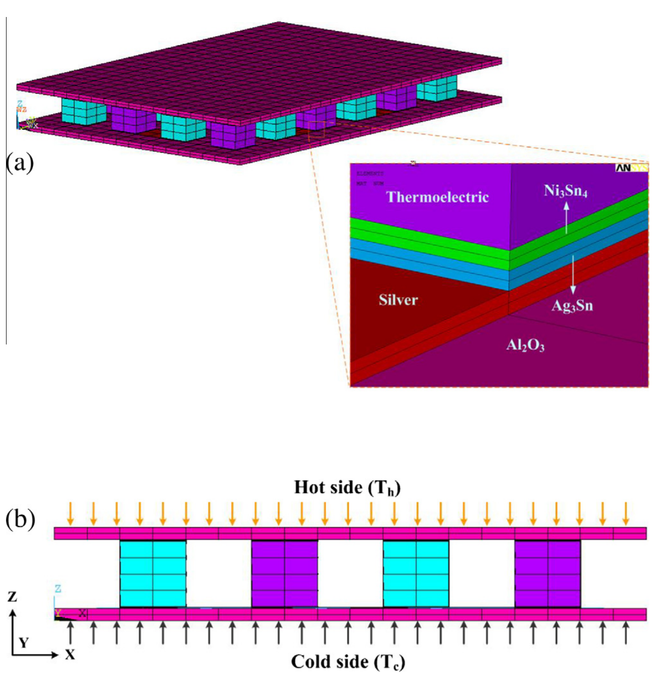

Fig. 4. (a) Established finite element model and (b) boundary condition of TEG.

3. Finite element model and material properties

Fig. 4(a) shows the established FE model of the TEG, which is based on an actual TEG. The TEG consists of 12 pairs of TE pillars, silver, Ag_3Sn IMC, Ni_3Sn_4 IMC, and ceramic substrates. The dimension of the TE pillars is 2 × 2 × 2 (mm^3). The thickness of silver, Ag_3Sn IMC, and Ni_3Sn_4 IMC are 18, 10.8, and 4.5 μm, respectively. The thermo-electrical behavior of the TEG with full IMC joints was predicted through thermo-electrical FE analysis with the commercial software ANSYS. Different temperature loads were set and applied on both sides of the TEG as the boundary condition (Fig. 4(b)). The temperatures on the hot and cold sides are 200 °C and 25 °C, respectively. The thermal load generated a temperature gradient in each TE pillar, and the electrical field was induced inside the materials. The material properties applied in the thermo-electrical FE analysis are listed in Table 1. The Seebeck coefficient of the TE material [9] affects the direction of the voltage produced in the circuit. Electrical resistivity and thermal conductivity influence the output power and temperature distribution of the TEG. The other material properties were set constant. The temperature distribution and output voltage of the TEG with full IMC joints were estimated at predicted thermal loads through numerical FE analysis.

4. Results and discussion

4.1. Joint test

Fig. 5(a) presents a cross-section image of the joint where both sides of tin react with silver and nickel to form Ag_3Sn and Ni_3Sn_4 IMCs after SLID bonding. The materials composition of the IMCs were detected and analyzed through EDS. The growth thickness of Ag_3Sn and Ni_3Sn_4 IMCs are 3.33 and 1.07 μm, respectively. For full IMC formation, the tin was entirely consumed and reacted with silver and nickel at a thermal treatment temperature of 200 °C. Fig. 5(b) presents the full IMCs formed in the joints after 3 d thermal treatment. Tin reacted completely with silver and nickel to transform into full IMCs. Only the Ni_3Sn_4 and Ag_3Sn IMCs remained at the joints. Fig. 6 shows the relationship between thermal treatment time and the growth thickness of the IMCs. Increasing the thermal treatment time increases the thickness of the IMCs. Thermal treatment time increases and the growth ratio of IMCs thickness decreases gradually in the solid–solid interdiffusion reaction.

4.2. Shear test

The mechanical strength of the joint was examined through a shear test at different thermal treatment times. The shear force of the joints was measured, and shear strength was calculated with the shear strength equation. Fig. 7 shows the relationship among thermal treatment time, shear force, and shear strength. The results reveal that increasing the thermal treatment time reduces the shear strength of the Ag_3Sn IMCs. In an ambient environment, the shear and tensile strength of the Ag_3Sn IMCs are lower than those of traditional lead-free solders [10,11] because IMC is a brittle material. However, the advantage of the Ag_3Sn IMC is that it has a melting point of 480 °C based on the phase image of Ag–Sn [12]. IMCs with a high melting point are applied as interconnection joints in TEGs and are allowed to operate at application temperatures above 200 °C. After the shear teat, the fracture surface of the joint was examined and analyzed through SEM and EDS. Fig. 8(a) and (b) shows the fracture surface of the joints after thermal treatment of 12 and 48 h, respectively. The major fracture occurred in the Ag_3Sn IMCs.

Table 1
Material properties applied in the thermo-electrical FE analysis.

	Thermal conductivity (W/mK)	Electrical resistivity (ohm m)	Seebeck coefficient (μV/K)
N-type (BiTeSe) [9]	1.6	9.8e-6	−170
P-type (BiTeSb) [9]	1.6	9.8e-6	170
Ni_3Sn_4	19.6	2.86e-7	–
Ag_3Sn	44.48	2.15e-7	–
Silver	419	1.55e-8	–
Al_2O_3	25	–	–

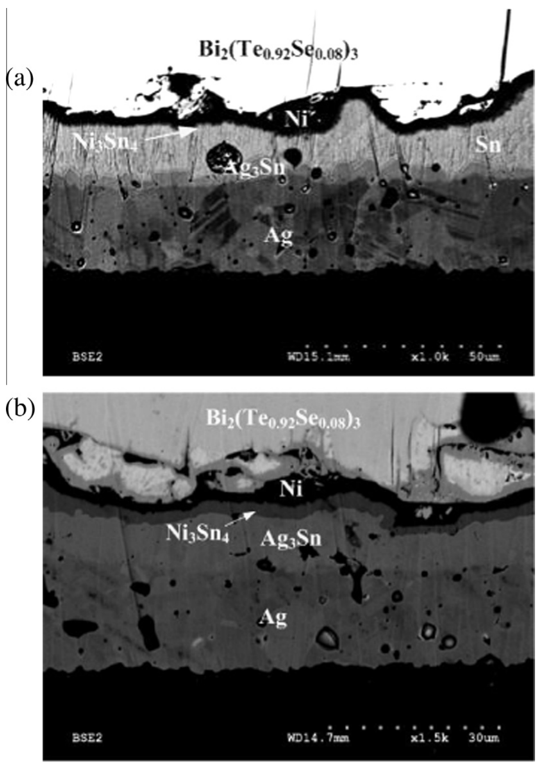

Fig. 5. Cross section image of the joint (a) after SLID bonding and (b) after 3 d thermal treatment.

4.3. Thermo-electrical analysis

With the established FE model of the TEG, thermo-electrical behavior was predicted at different temperature loads through numerical FE analysis. The temperature load on the hot and cold sides is 200 °C and 25 °C, respectively. Temperature load generates a temperature gradient in the TE pillar and induces potential voltage outputting inside the TEG. Fig. 9 shows the temperature gradient distribution of the TEG formed at a temperature difference of 175 °C. The temperature of the TEG declines from the hot side to the cold side. The thermal conductivity of the materials is the major factor that caused the temperature gradient. The Seebeck coefficient and the electrical resistivity of the material also affect the generation of the potential voltage. Fig. 10 shows the potential voltage of the TEG predicted at a different temperature of 175 °C with a potential voltage of 0.31 V. The temperature distribution and potential voltage of the TEG with full IMC joints were rapidly estimated and predicted at a predetermined temperature load through numerical analysis.

4.4. TEG assembly and performance measurement

Ni/Sn/Ag was utilized as the interconnection joint assembled with the TE pillars to form the TEG. The 12 pairs of TE pillars are shown in Fig. 11. A short circuit may occur at the joints because tin is thin and does not fill the gap between the TE pillar and the substrate. Therefore, a TEG with three pairs of TE pillars was fabricated to measure the TE characteristics. The Seebeck coefficient,

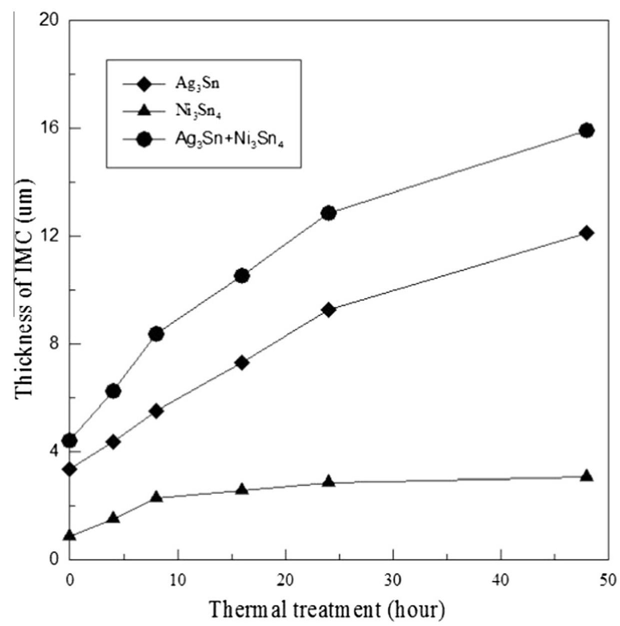

Fig. 6. Relationship between the thermal treatment time and the growth thickness of IMCs.

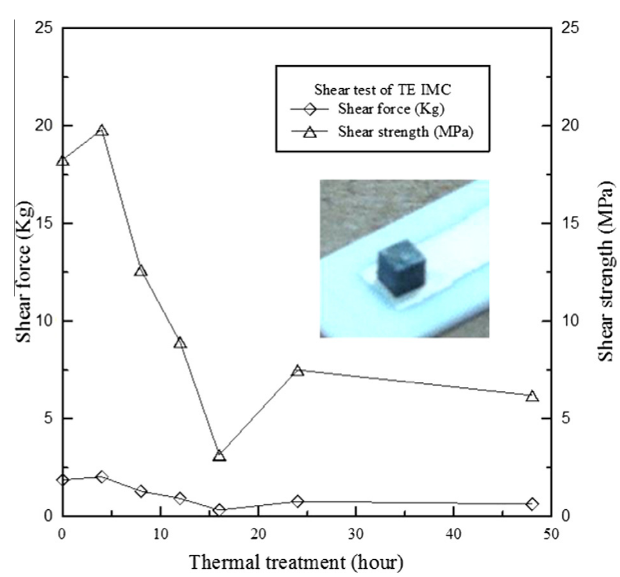

Fig. 7. Relationship among thermal treatment times, shear force and shear strength.

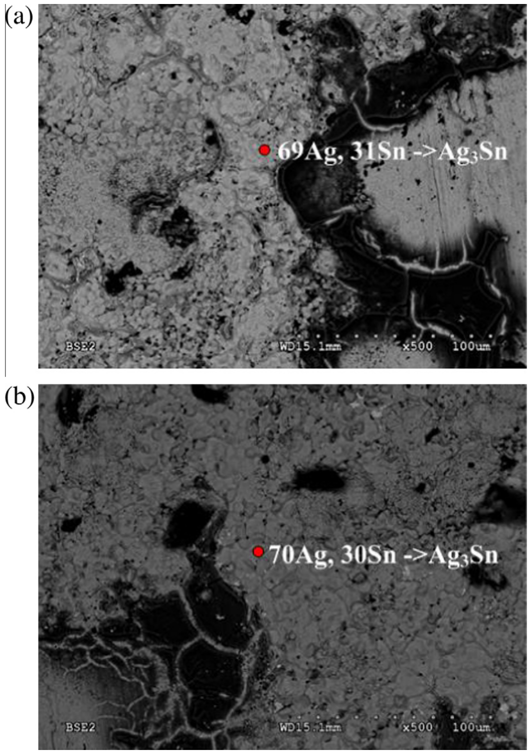

Fig. 8. Fracture surface of joints after thermal treatment of (a) 12 and (b) 48 h.

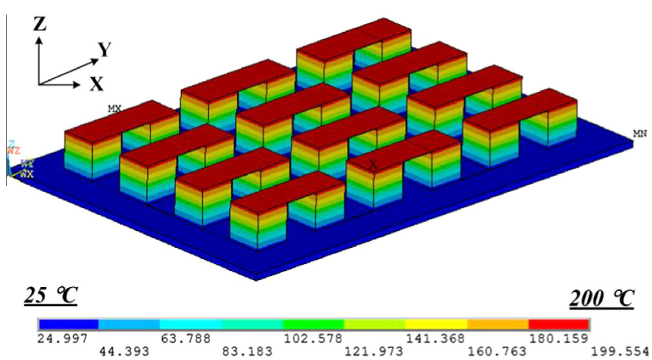

Fig. 9. Temperature gradient distribution of the TEG formed at a temperature difference of 175 °C.

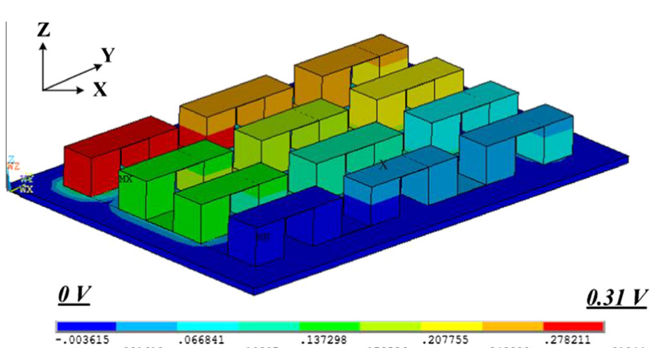

Fig. 10. Potential voltage of the TEG predicted at temperature difference of 175 °C with a potential voltage of 0.31 V.

electrical resistance, and thermal conductivity of the TE material and module were measured through the slope method [13]. The measurements of the characteristics of the TEG are shown in Table 2. The results show that the characteristics of TEG do not vary significantly before and after thermal treatment, that is, IMC as the interconnection joint has a minimal effect on the performance of the TEG. The high melting point of IMC allows the TEG to operate at temperatures above 200 °C.

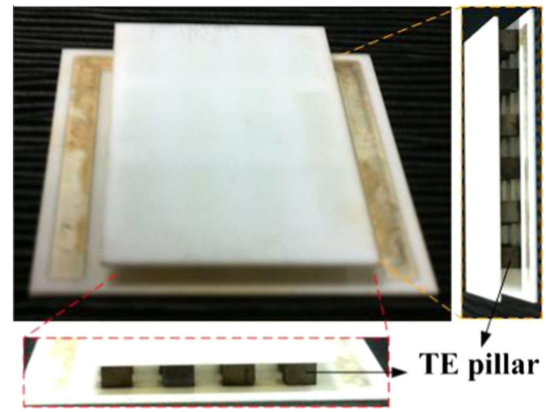

Fig. 11. Ni/Sn/Ag was utilized as the interconnection joint assembled with the TE pillars to form the TEG, which has 12 pairs of TE pillars.

Table 2
TE characteristic of the TEG.

	Seebeck coefficient (μV/K)	Electrical resistance (ohm m)	Thermal conductivity (W/mK)
Materials	181.74	$1.64e^{-5}$	2.26
Materials (Thermal treatment)	192.25	$1.61e^{-5}$	2.65
Module	173.90	$1.65e^{-5}$	2.14
Module (Thermal treatment)	177.26	$1.63e^{-5}$	2.44

5. Conclusion

In this study, Ni/Sn/Ag was adopted as the joint material and applied in the TEG. Tin reacted with silver and nickel to form Ag_3Sn and Ni_3Sn_4 IMCs after SLID bonding. Tin was entirely consumed and reacted with silver and nickel to transform into a full IMC through solid–solid interdiffusion reaction. Only the Ag_3Sn and Ni_3Sn_4 IMCs remained at the joint. The mechanical strength of the joints was examined through a shear test at different thermal treatment times. An increase in thermal treatment time results in an increase in the thickness of IMC and a decline in the shear strength of the joint. Ni/Sn/Ag was utilized as the interconnection joint to be assembled with the TE pillar to form the TEG. The characteristics of the TEG do not vary significantly before and after thermal treatment. The findings reveal that IMC exists on the joints and has a minimal effect on the performance of the TEG. An IMC with a high melting point was used as the joint and applied in the TEG, which can operate at temperatures above 200 °C. The thermo-electrical behavior of the TEG, such as temperature distribution and potential voltage, was estimated at a specific temperature load through numerical FE analysis. Thermo-electrical FE analysis can help us rapidly estimate and predict the performance of the TEG at a predetermined temperature load.

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